INVESTIGATION OF LASER DYNAMICS, MODULATION AND CONTROL

BY MEANS OF INTRA-CAVITY TIME VARYING PERTURBATION

under the direction of

S. E. Harris

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INTRODUCTION

The work under this grant is generally concerned with the generation, control, and stabilization of optical frequency radiation. In particular, we are now nearly exclusively concerned with attaining tunable optical sources by means of nonlinear optical techniques. The present projects supported by this grant include: (a) an experimental and theoretical study of a new type of optical radiation which we have termed as parametric fluorescence or spontaneous emission; (b) a project aimed at the attainment of a parametric oscillator employing a stable, phase-locked gas laser pumping source; and (c) a project whose goal is the demonstration of backward wave parametric oscillation wherein optical resonators need not be employed, and wherein the output should be tunable over a large region of the far infrared spectrum.

During this period the following publication has been submitted for publication.

R. L. Byer and S. E. Harris, "Power and Bandwidth of Spontaneous Parametric Emission," to be published in Physical Review.

The following oral disclosures have also been presented:

S. E. Harris, R. L. Byer, and M. K. Oshman, "Power and Bandwidth of Optical Parametric Fluorescence," third national Conference on Nonlinear Optics, Erevan, Armenia, USSR, October 1967.

PRESENT STATUS

1(a). Optical Parametric Fluorescence (R. L. Byer and S. E. Harris)

The parametric fluorescence theory-experiment check has been completed, and a paper has been accepted for publication in Physical Review. The results are presented below.

A theoretical temperature tuning curve was generated on the computer using the data of Hobden and Warner. Figure 1 shows that the experiment and theory agree well. We found that although the slope of the curve was the same for all crystals measured, some curves were displaced in temperature by as much as $30^{\circ}C$ from the theory. This difference can be attributed to differences in crystal indices due to crystal impurities and inhomogeneities.

The predicted linear relationship between fluorescence power and acceptance angle squared is compared to theory in Fig. 2. The data points indicate that the experimental value of the lithium niobate nonlinearity, $d_{15} = 0.47 \times 10^{-22}$ mks, is slightly less than the published value of 0.56×10^{-22} mks. The measurement of crystal nonlinearity is an important application of parametric fluorescence. Two important advantages of this method relative to second harmonic generation are first, that the measurement is independent of the area of the pumping beam, and second that the result is dependent only on the average power of the pumping laser and independent of the commonly occurring temporal fluctuations.

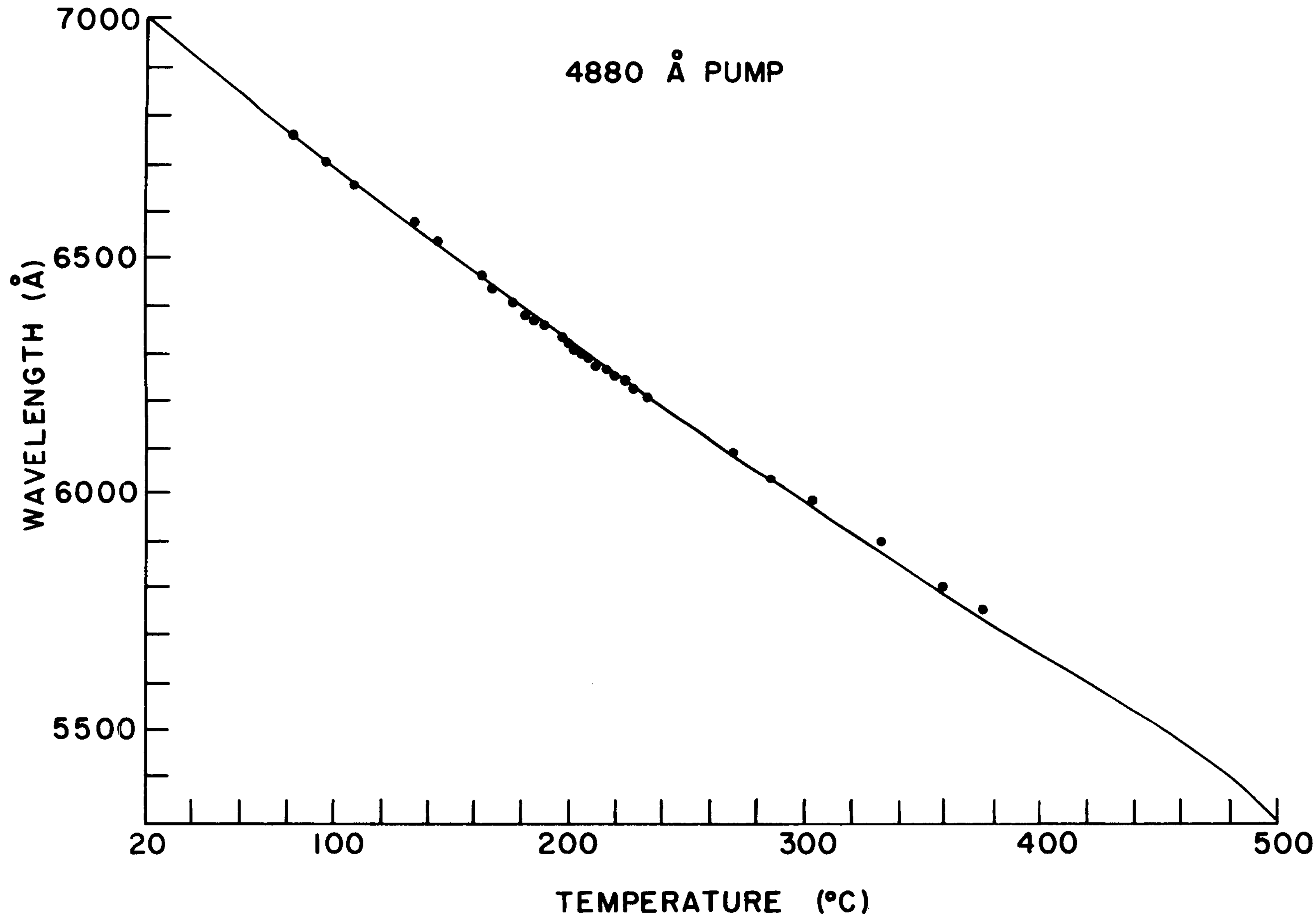

FIG. 1--Signal wavelength vs crystal temperature, theoretical curve and measured points.

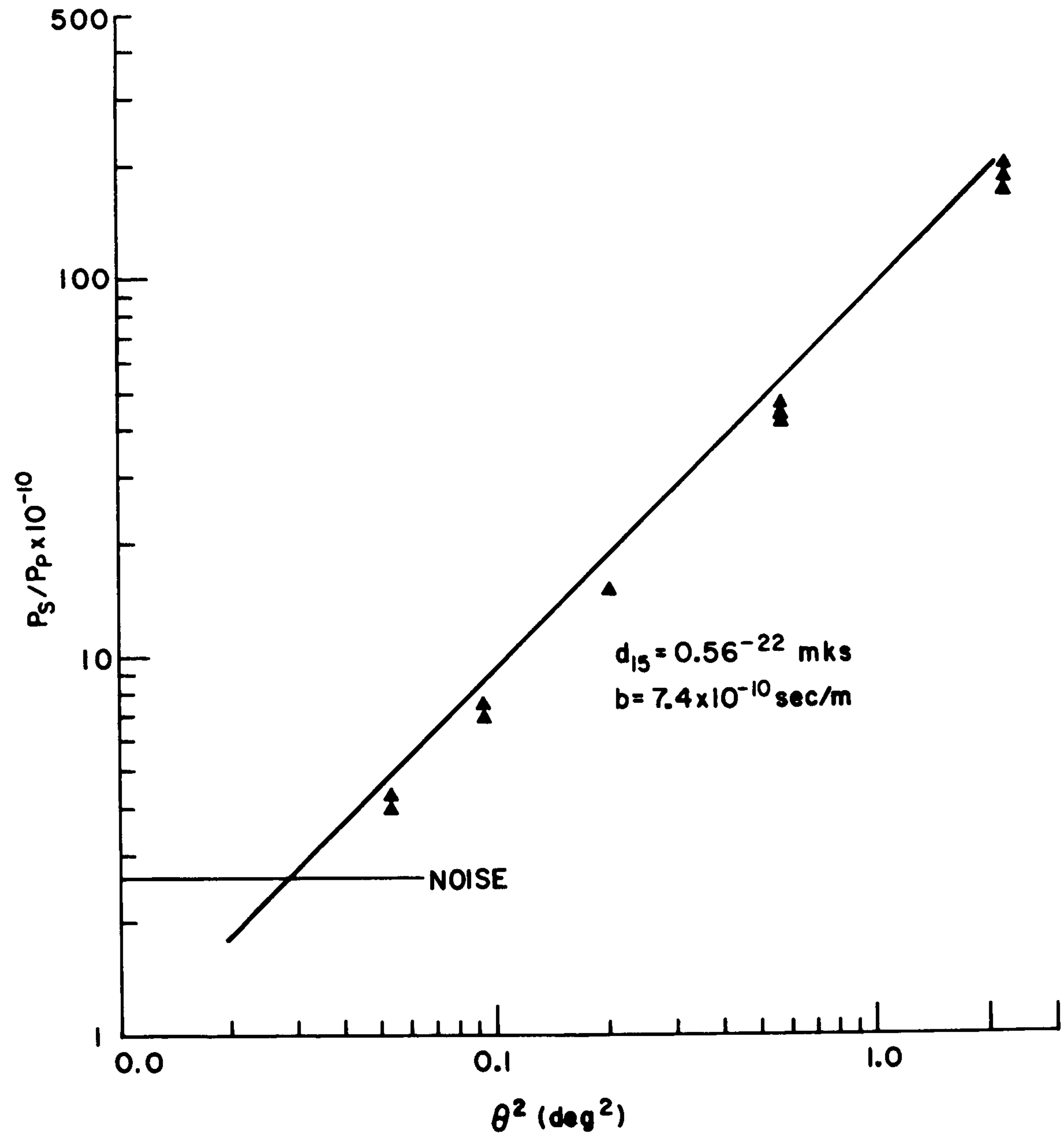

FIG. 2--Total spontaneously emitted power vs Θ^2 showing theoretical and experimental results.

Theoretical curves of fluorescence power as a function of frequency for various acceptance angles are shown in Fig. 3. The experimental points shown in part (c) verify the theory very closely. The bandwidth versus θ^2 curve of Fig. 4 presents the data in a more graphic form. This curve shows that the bandwidth approaches a predicted minimum value for small angles. The quantitative disagreement of experiment and theory is believed to be due to crystal impurities which effect the indices.

At present parametric fluorescence is proving very valuable for determining the tuning curves and nonlinearity of crystals to be used in the parametric oscillator experiments.

1(b). <u>Fluorescence and Superradiance in $LiNbO_3$ Pumped at $\lambda_p = 0.694\mu$</u>

(J. Falk, J. E. Murray, S. E. Harris)

The possibility of phase matching a 3-frequency down conversion parametric interaction in $LiNbO_3$ with a pump at 0.694μ has been pointed out by Hobden and Warner.[1] During this past period we have experimentally verified this direct phase matching and obtained the portion of the phase matching curve between 0.8μ and 1.0μ. Also we have attempted to experimentally demonstrate parametric gains on the order of 30 dB with this process. Unfortunately, material problems have made the results inconclusive.

The primary importance of this process is that it potentially provides very high gain which is tunable from about 0.8μ to 4.0μ. This is possible for the direct phase matched process because it does not

[1] J. Warner and M. V. Hobden, Phys. Letters 22, 243 (August 15, 1966).

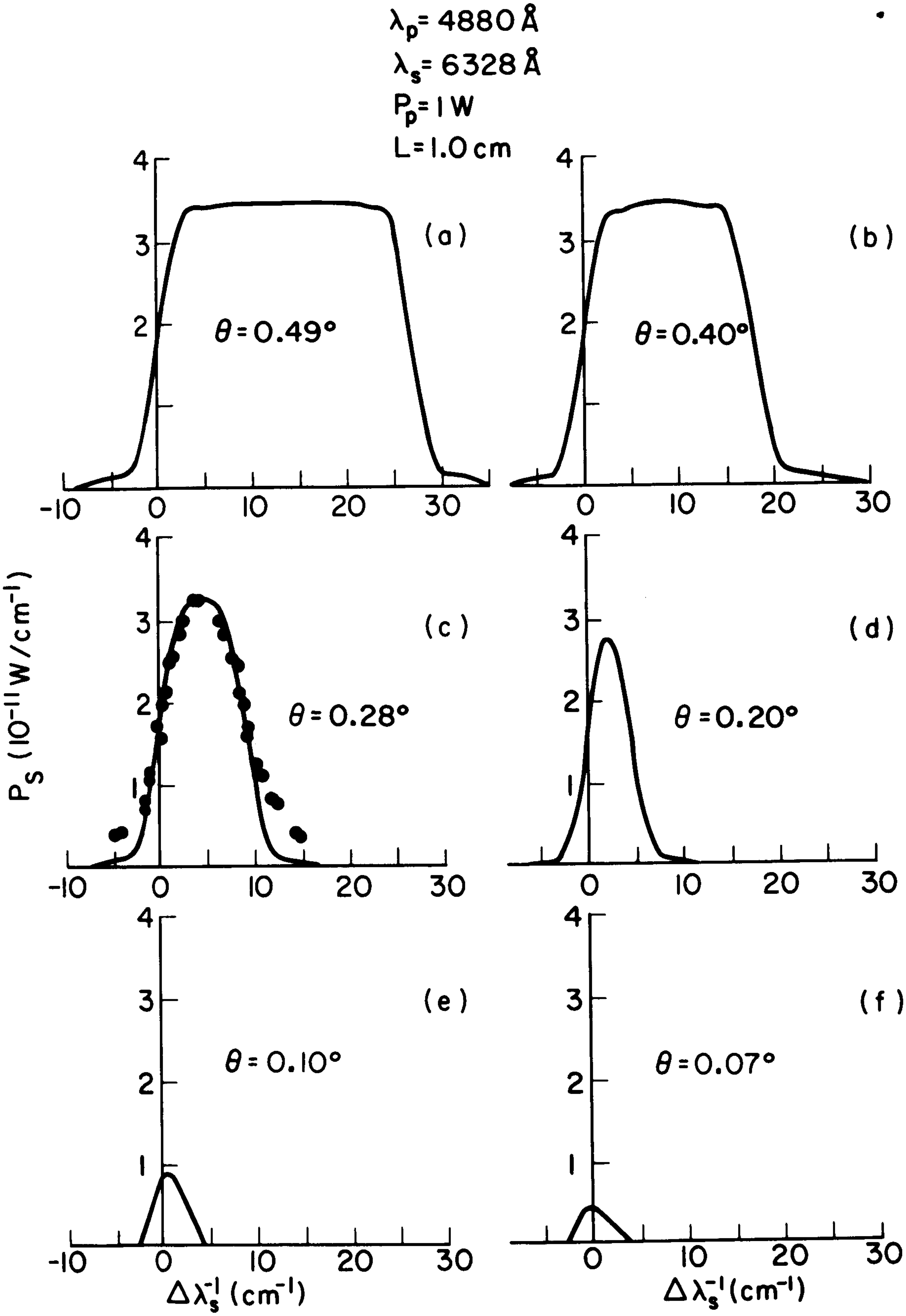

FIG. 3--Spectral distribution of spontaneous power at different acceptance angles. Part (c) shows experimental points normalized to peak of the theoretical curve.

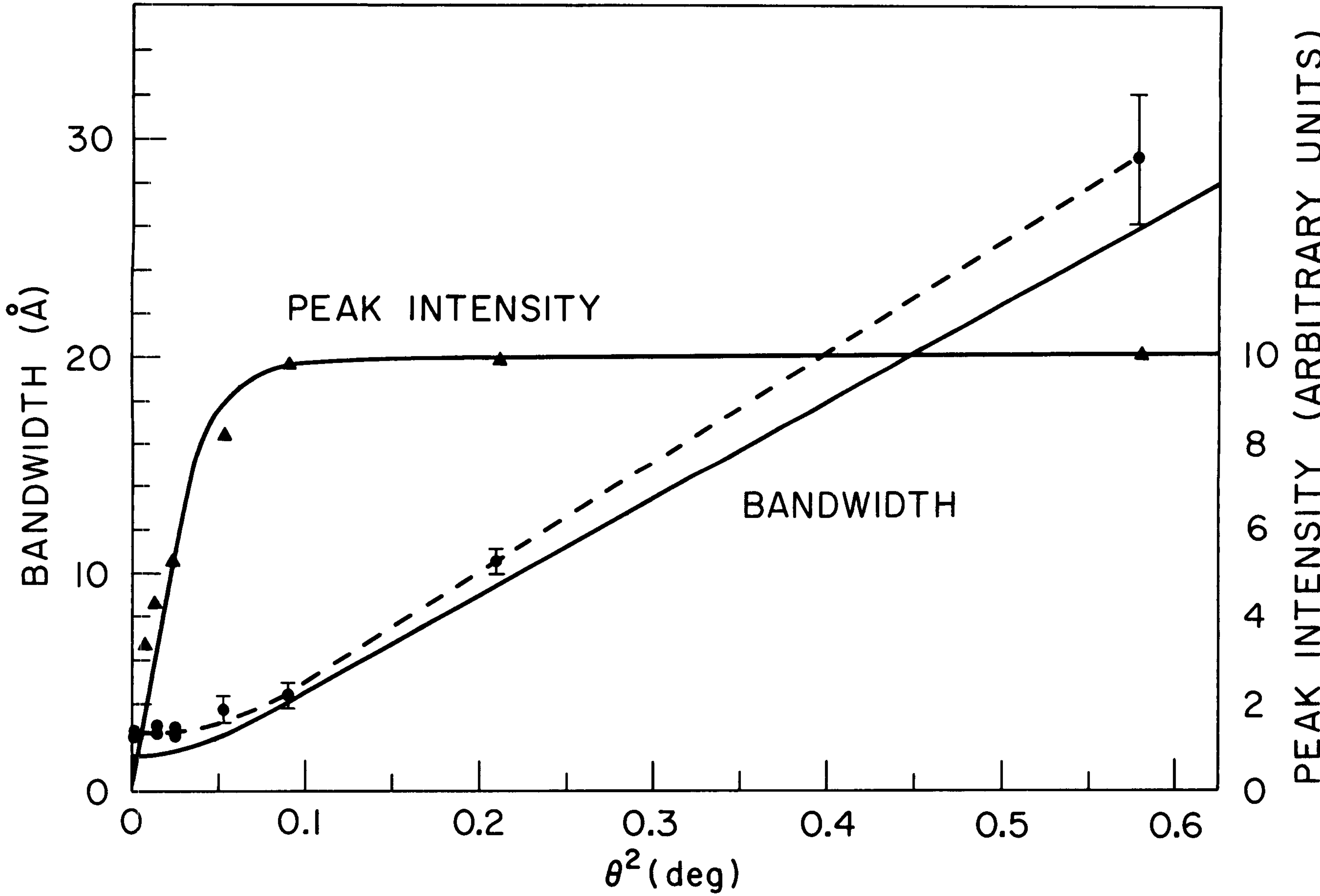

FIG. 4--Peak intensity and bandwidth vs acceptance angles. Solid line is theoretical and the dotted line is experimental.

require frequency doubling of the pump and thus makes possible much higher pump densities. These high gains would, for example, make it possible to achieve oscillation threshold with very wide band but lossy cavity mirrors, giving a source which would be tunable over a large portion of the near infrared spectrum. Also, high gains would eliminate the need for resonating both the signal and idler, thus avoiding the frequency hopping characteristic of cavities which resonate both.[2,3] Also of importance is the possibility of using this process in a tunable infrared amplifier.

The first experimental objective was to determine the tuning curve for the 3-frequency down conversion process. The phase matching conditions are the usual ones for such a process:

$$\omega_p = \omega_s + \omega_i$$

$$k_p = k_s + k_i \quad ,$$

where the subscripts p , i , and s refer to the pump, idler, and signal modes, respectively. We will define the signal as that mode which tunes from degenerate at 1.3μ to shorter wavelengths, because this is the mode we detected experimentally. Tuning is achieved by means of the temperature dependence of the refractive indices of the $LiNbO_3$ crystal.

Using the data of Hobden and Warner, we obtained the theoretical tuning curve shown in Fig. 5 from a computer program. This curve guided

[2] J. A. Giordmaine and R. C. Miller, Phys. Rev. Letters 14, 973 (June 14, 1965).

[3] J. A. Giordmaine and R. C. Miller, Appl. Phys. Letters 9, 298 (1966).

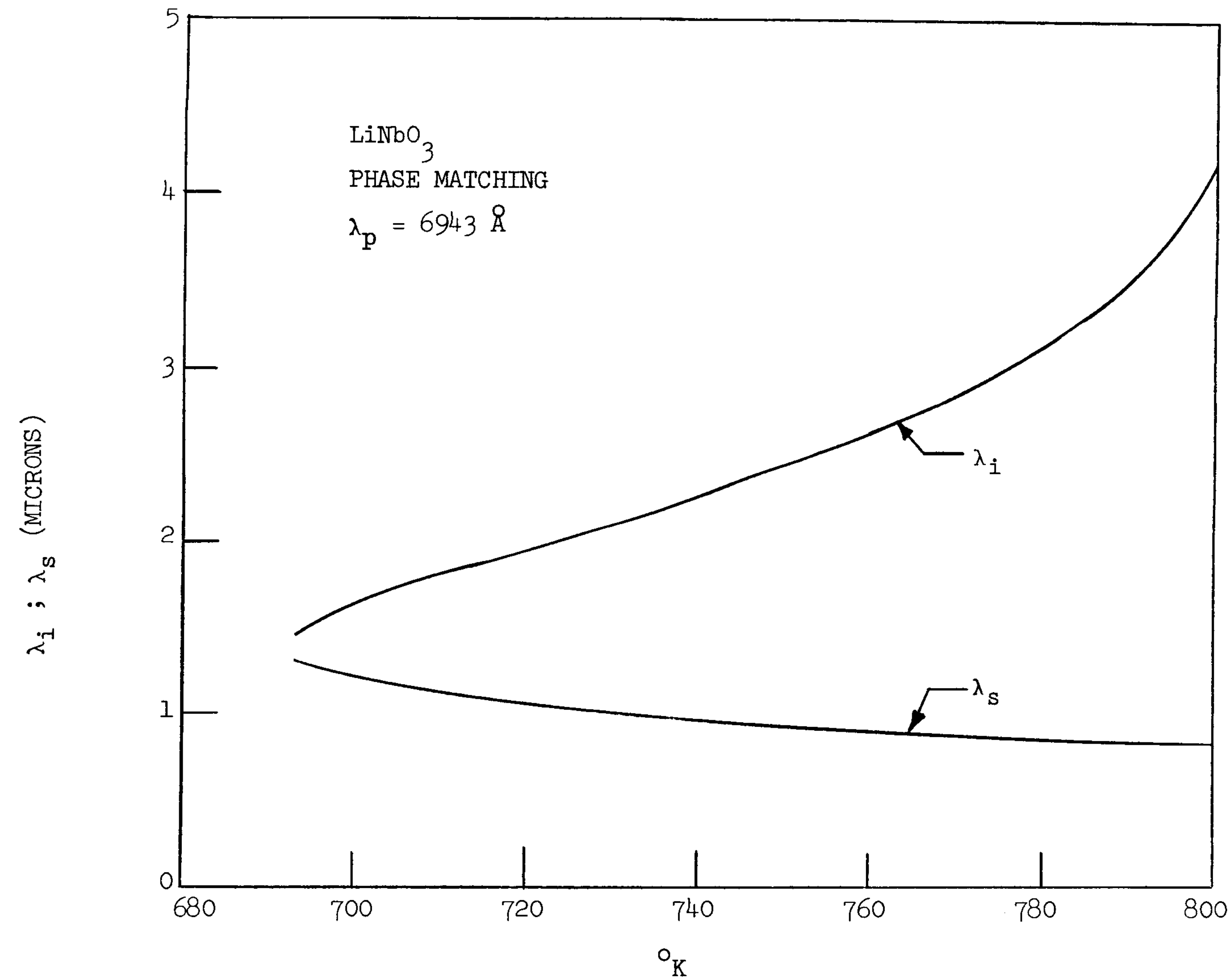

FIG. 5--Theoretical tuning curve, lithium niobate pumped with ruby.

us to the general temperature range for phase matching. Then, by observing fluorescent emission produced by pumping with about 10 kW/cm^2 we obtained the tuning curve shown in Fig. 6. We found the actual phase matching temperatures to be about 40°K higher than those predicted by theory. The wavelength of the emission was determined with a Zeiss Monochrometer followed by a DuMont photomultiplier with an S-1 surface. The monochrometer slits were set to give a resolution of about 0.03μ which was a compromise between ease in locating the signal and accuracy of the resulting phase matching curve. The experimental result of Fig. 6 also shows the width of the observed detector response, indicated by the solid lines at the data points. For the 5 data points below 825°K, this apparent wavelength spread is due to the monochrometer resolution; but the points above 825°K indicate a very appreciable frequency broadening in the emission as the idler begins to enter the lossy region at about 4.0μ. This is predicted by the theoretical bandwidth for fluorescent emission as given by Byer and Harris.[4]

The next experimental objective was to demonstrate the large gains predicted by theory. The procedure used was to monitor the signal power, which resulted from single pass gain of spontaneous signal power, as the pump power density was increased. Thus, we expected to see an exponential increase in signal power with a linear increase in pump power density.

An extension of the fluorescent theory of Byer and Harris[4] resulted in the following expression for signal power density as a function of

[4] R. L. Byer and S. E. Harris, Microwave Laboratory Report No. 1595, Stanford University (November 1967); to be published in Phys. Rev.

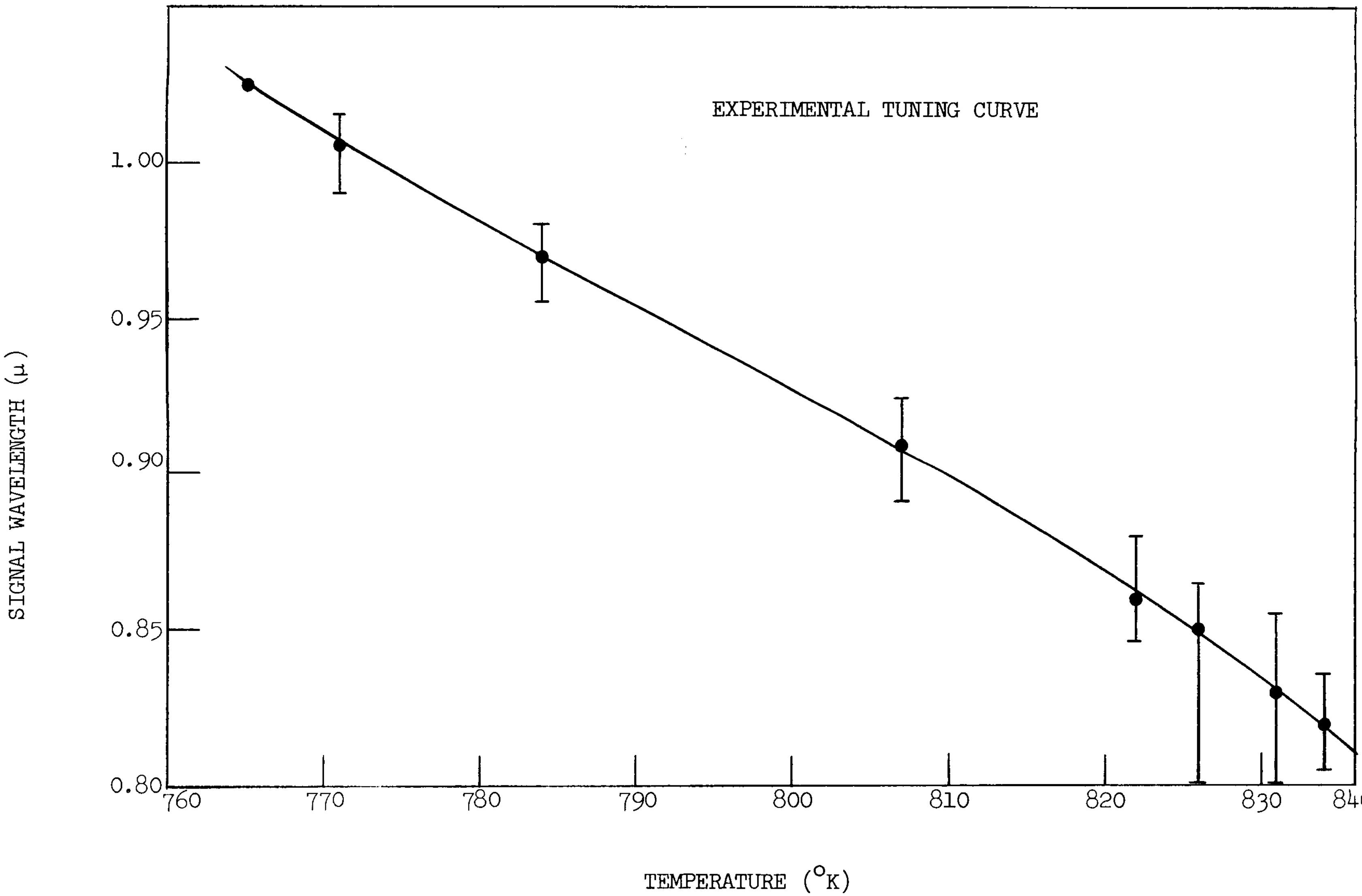

FIG. 6--Experimentally determined tuning curve. Solid lines indicate width of observed detector response.

beam radius:

$$I_s(r) = \frac{\hbar n_s^2 \omega_s^3}{4\pi^2 \, bLc^2} (\Theta^2)[L\Gamma(r)]$$

where

$$b \triangleq \left[\frac{\partial k_i}{\partial \omega_i} - \frac{\partial k_s}{\partial \omega_s} \right]$$

Θ = polar acceptance angle in the crystal

L = crystal length

n_s = refractive index of signal

$$\Gamma(r) = \sqrt{\frac{\omega_s \, \omega_i}{n_s^3 \, c^3 \, \epsilon_0^3} \left(\frac{d^2}{2}\right)} \sqrt{I_p(r)}$$

d = electro-optic coupling constant.

Since we measured total power, it was necessary to integrate the signal power density over the assumed Gaussian cross section of the pump:

$$P_s = (2\pi) \frac{\hbar \, \omega_s^3 \, n_s^3 \, \Theta^2}{8\pi^2 \, b \, L} \int_{\infty}^{\infty} \sinh^2 [L\Gamma(r)] \, rdr \quad .$$

The near field for our setup was a factor of 50 times longer than our crystal so we neglected the variation of pump density along the beam axis. Making a change of variable in the above integral, we obtain

$$P_s = B \, A \int_0^a \left(\frac{\cosh \, u - 1}{u} \right) du \quad ,$$

where B is the collection of constants preceding the integral in the previous expression; A is the pump beam area using its Gaussian waist; and

$$a = 2\sqrt{2}\, L\, \Gamma(0)$$

with $I(0)$ given by the total pump power divided by its Gaussian area.

Figure 7 shows the experimental results for one of our $LiNbO_3$ crystals. For this particular crystal, the effective electro-optic constant was measured to be

$$d = 0.81 \times 10^{-22} \text{ (mks)} \quad ,$$

by SHG from $\lambda_p = 1.15\mu$. The crystal length was 1.5 cm. Also shown on the figure are the exponential responses predicted by the above equation and the linear response for this crystal.

As can be seen from the figure, only a few of the experimental points are consistent with the expected exponential response. The upper points indicate a gain of about 27 dB. Most of the others are on or below the theoretical linear response.

We attribute the points, which seem to indicate a linear response, to crystal damage. We found that we scorched the surface of our crystal with 60 - 80 MW/cm^2, and that subsequent shots at this same spot would invariably give proportionally smaller signals. This is consistent with the physics of the interaction because the damaged areas of the crystal would act to diffuse the beam, destroying the plane wave character of it by distributing its power throughout a large solid angle. Since this interaction will accept only that pump power within

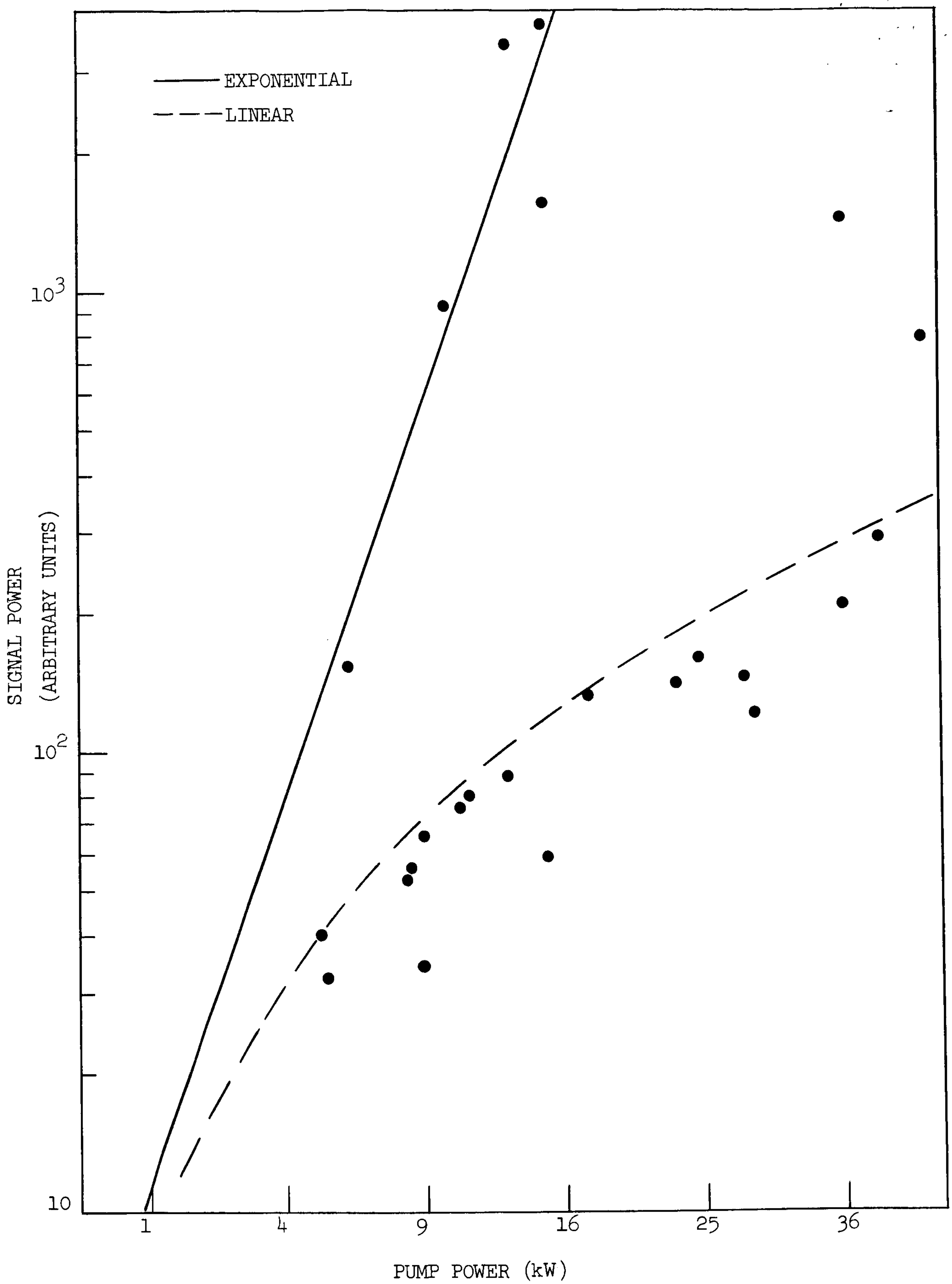

FIG. 7--Fluorescent power output as a function of pump power. Solid line indicates theoretical power output. Dashed line indicates linear extrapolation of theoretical fluorescent power output.

a limited polar angle to pump a single signal mode, this diffuse scattering would greatly reduce the effective pump power per signal mode. Also, our detecting system had a reasonably narrow angular acceptance, $\theta_{max} = 2.24$ milliradians, which would have prevented us from seeing off-angle signal modes.

In conclusion, our data is encouraging but not by any means decisive. We are hoping to acquire a good crystal of the new nonlinear material $Ba_2NaNb_5O_{15}$. With this crystal we can achieve the same gain levels with only one-ninth of the pump power density required in $LiNbO_3$. Thus we hope to avoid the damage problem we have encountered thus far.

It is interesting to note that with sufficiently good crystals of $Ba_2NaNb_5O_{15}$ it would be possible to reach gain levels high enough to amplify spontaneous noise power to useful levels, resulting in a tunable superradiant source.

2. Parametric Oscillation at Optical Frequencies (R. L. Byer, J. F. Young, and S. E. Harris)

Components for the internal argon pumped parametric oscillator described in the previous report have been completed and tested. The experiment was not successful because of the excess cavity loss introduced by the $LiNbO_3$ within the laser cavity. Figure 8 shows the measured absorption loss of $LiNbO_3$. Power absorbed at the laser pumping wavelength causes thermal self-focusing. The crystal locally heats and becomes a high power positive lens which disrupts the cavity mode and makes it extremely difficult to obtain high power in the fundamental Gaussian mode. The best observed laser power at 4800 Å

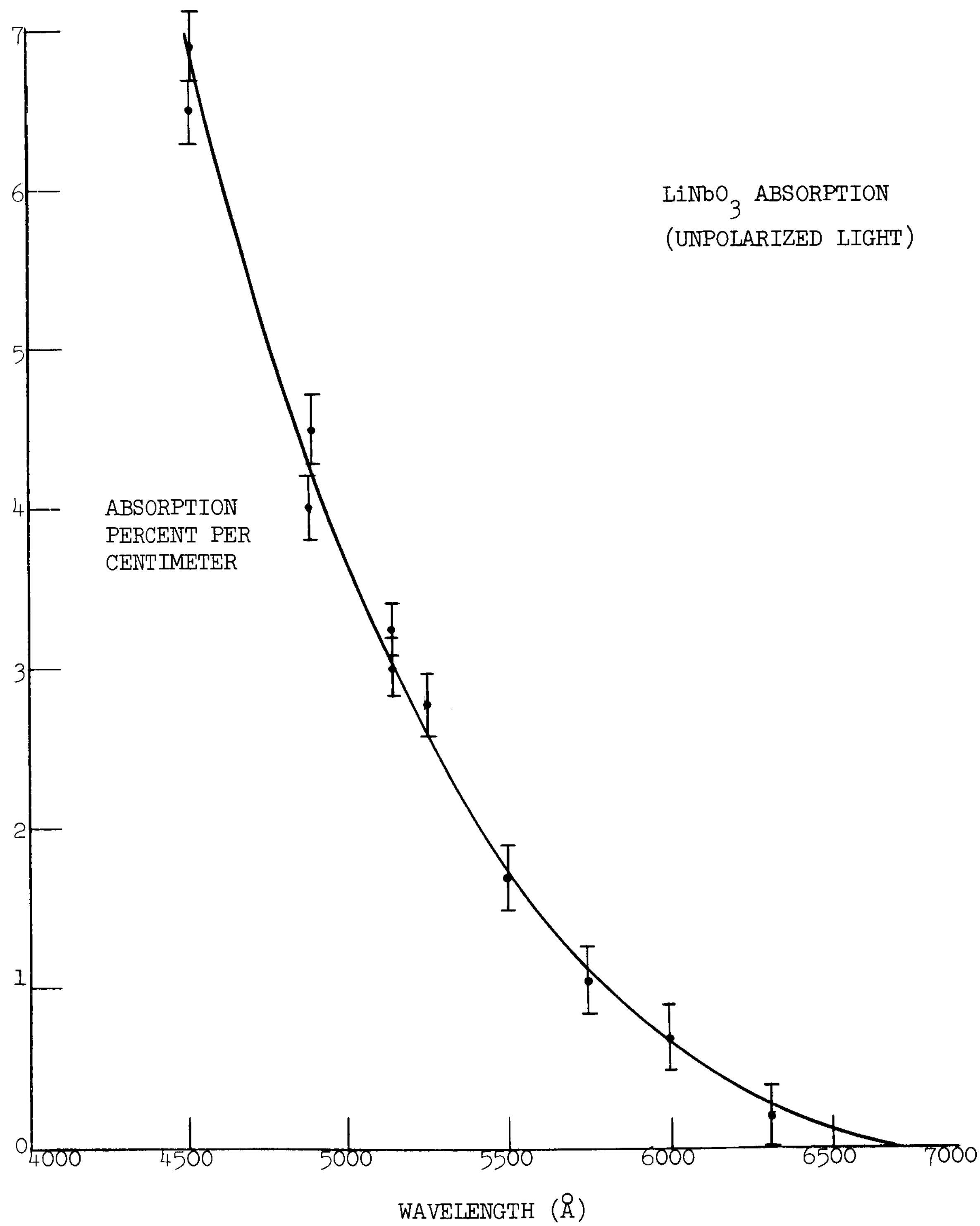

FIG. 8--Absorption loss in $LiNbO_3$ as a function of wavelength.

was 1.5 watts, well below the expected 4-5 watts. The beam splitter developed for this internal oscillator was successfully coated and assembled.

In the last report we described the failure of the initial krypton-pumped external parametric oscillator because of poor crystal qualities. Since then we have made considerable progress in the growing, poling, and testing of $LiNbO_3$ crystals. The crystals are grown by the Material Science Department at Stanford and poled by us to produce crystals having a single ferroelectric domain. After poling, four tests are used to verify crystal quality: usual inspection between crossed polarizers for strain; measurement of electro-optic half-wave voltage; measurement of nonlinear coefficient d_{15} by SHG; and measurement of the temperature tuning curve using parametric fluorescence, which also provides an independent measurement of d_{15} .

All six crystals which were poled are strain free, but only three have satisfactory half-wave voltages. A poor half-wave voltage is believed to indicate incomplete poling (multi-domain crystal) and/or large impurity concentrations. The SHG test is made by slowly varying crystal temperature while recording second harmonic power output. Theoretically this power is proportional to $(\sin k\Delta T/|k\Delta T|)^2$ where k is a constant and ΔT is the temperature deviation from the exact phase-matching temperature. Figure 9 shows a SHG curve which agrees very well with the theory. Second harmonic power is generated only over a very narrow temperature range, and the value of d_{15} computed from this data agrees closely with the accepted literature value. Figure 10 shows a curve typical of the other two crystals measured.

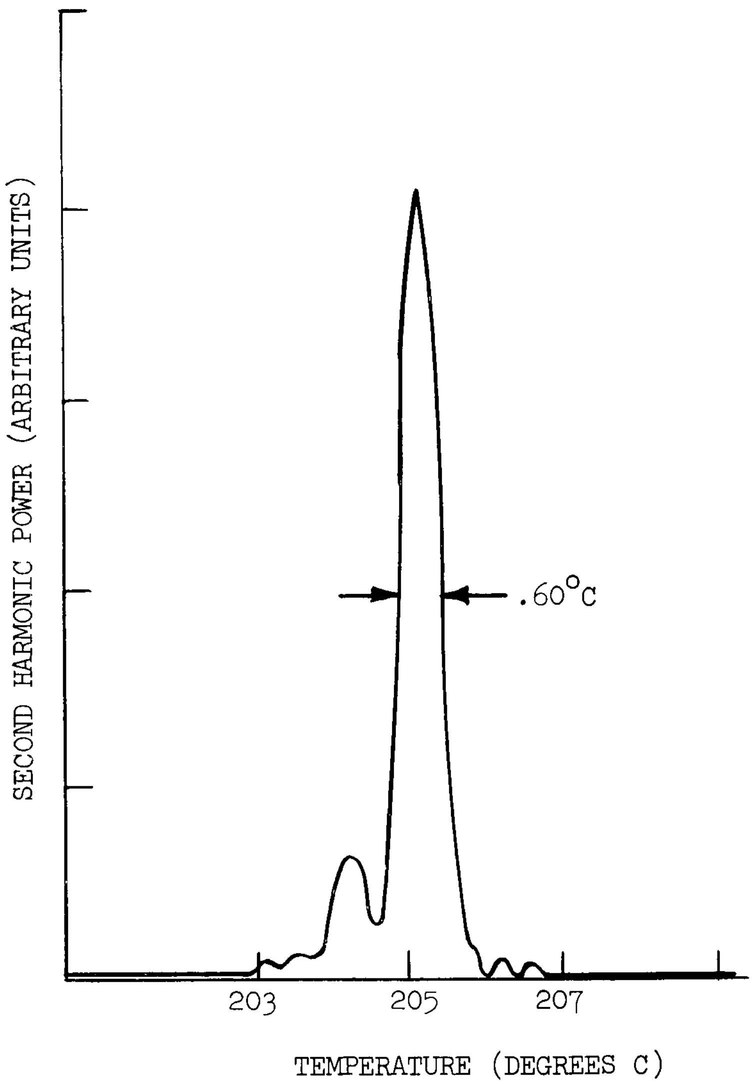

FIG. 9--Second harmonic generation (1.15 μ to 0.575 μ) of $LiNbO_3$ crystal No. 103, 1.7 cm long.

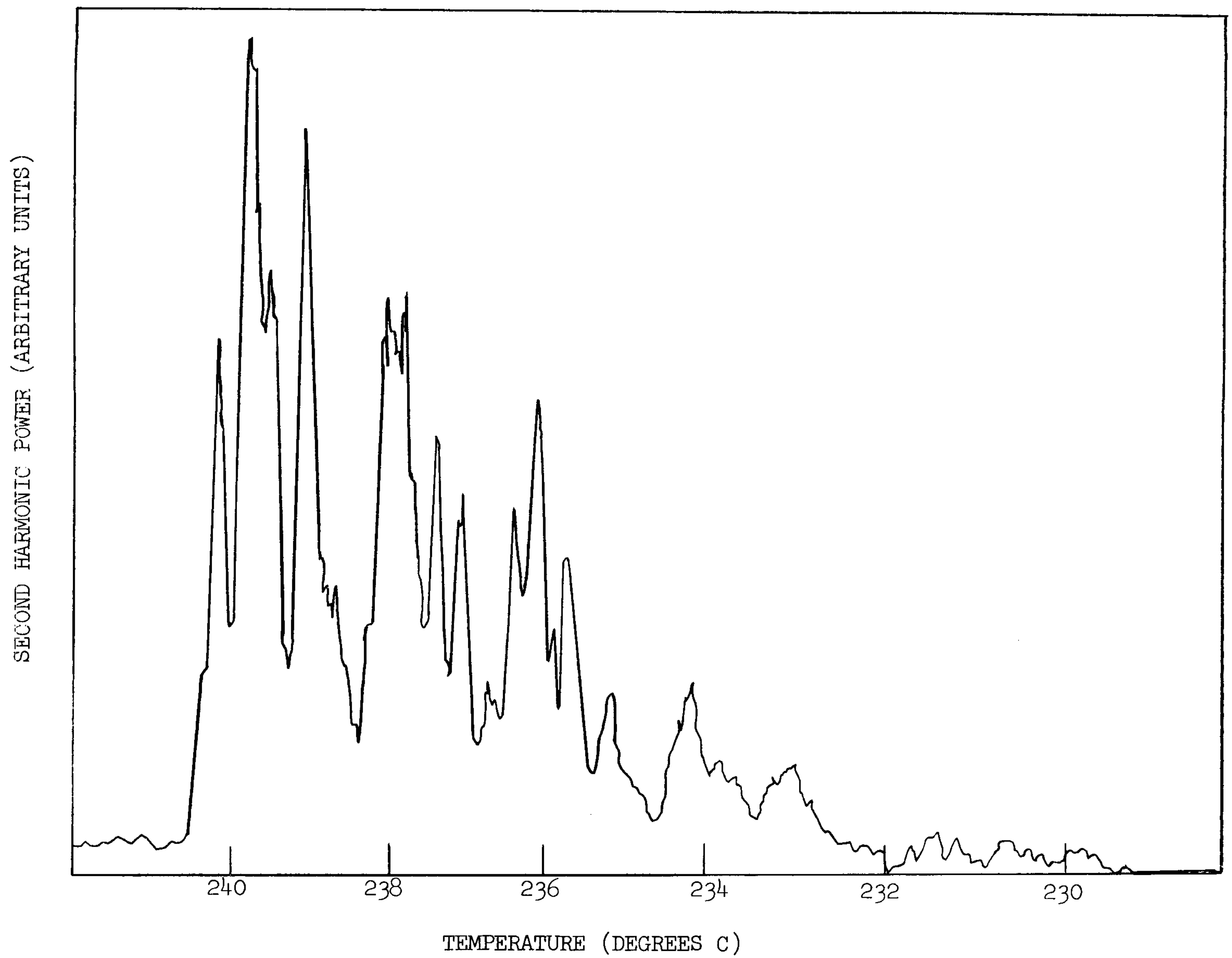

FIG. 10--Second harmonic generation (1.15 μ to 0.575 μ) of $LiNbO_3$ crystal No. 115, 2.4 cm long.

These longer crystals generate second harmonic power weakly over a wide temperature range and are therefore not useful. This type of crystal prevented the krypton experiment from working. At the present time the cause of this smearing is not known with certainty, but it is suspected to be due to inhomogeneous inclusion of impurities during growth. Long crystals are particularly susceptable to this difficulty and crystal #103 (Fig. 1) represents a 60% improvement in length over our previous best crystal. We are now testing crystals grown along a different axis which we hope will yield good crystals 2.5 to 3 cm long. Longer crystals and therefore greater parametric gain will increase the oscillation tuning range as well as ease all the critical tolerances in the system.

Considering the high quality of crystal #103 and the difficulties of the krypton experiment, we decided to attempt to construct an external parametric oscillator using the argon ion laser 4880 Å line as a pump. The argon laser is generally more stable than krypton and it is easier to obtain high pumping powers. In addition the argon plasma tubes last longer.

Degenerate phase matching with a 4880 Å pump is not possible with $LiNbO_3$. Therefore a unique operating point was chosen at which the cavity mirrors could be made highly reflecting at signal and idler wavelengths simultaneously. By picking the idler wavelength to be three times the signal wavelength, mirror and anti-reflection coatings can be made which are a quarter wavelength thick for the idler and three-quarter wavelengths thick at the signal — both highly reflecting.

The cavity was constructed and tested. The total effective (geometric mean) single pass loss at signal (6450 Å) and idler (2.00μ) was 3%. This was low enough that with available pump power the oscillator was above threshold. The experiment was tried but was not successful for two reasons: (1) The $LiNbO_3$ crystal thermally focused which decreased the power coupled into the cavity modes; and (2) we were unable to phase-lock the argon laser at 4880 Å sufficiently well because of the very high excess gain of that line. After evaluating these difficulties, we believe the argon line at 5145 Å will be a more suitable pump. Because of its lower gain, it can be phase-locked very strongly. This gain more than compensates for the small loss in output power relative to 4880 Å. The parametric cavity mirrors are presently being coated for this experiment. The thermal self-focusing effect will be somewhat reduced because of the lower crystal absorption at 5145 Å (Fig. 8). In addition the time average power into the $LiNbO_3$ will be reduced with a light chopper of suitable duty cycle.

Simultaneously with the experimental work, a theoretical analysis of the parametric oscillator with a phase-locked pump is being done. This will answer problems of mode competition, effective peak power available, and parametric cavity length tolerances. In addition, the problem of optimum pump focussing and cavity design is being studied.

3. Backward Wave Oscillation in the Far Infrared (J. Falk, J. E. Murray, and S. E. Harris)

The purpose of this project is to demonstrate a technique for obtaining large amounts of tunable far infrared power. The basic idea is to make use of a backward wave interaction of three electromagnetic waves which are coupled together by means of the electro-optic non-linearity.

The experiment attempted, as proposed in Status Report 2, makes use of a Q-switched ruby laser and a crystal of $LiTaO_3$. For sufficiently high pump densities optical cavities for signal and idler are not required and feedback is provided internally to the crystal. The calculated threshold pump density for the 2.5 cm crystal used in our experimental efforts was 12 Mw/cm^2. Several unsuccessful attempts were made at achieving oscillation. Pump densities of 70-125 Mw/cm^2, with pumping beam divergence of 10-20 milliradians were used.

Failure to attain oscillation threshold is attributed to laser beam divergence, i.e., it was not possible to focus the laser beam down to a spot size that would theoretically be above threshold and that would have a divergence that did not violate k-vector synchronism.

Subsequent to our experimental efforts an analysis was carried out to determine the allowable pump angular spread which could drive the parametric process.

The results of this analysis show that pump energy in a polar angle given by

$$\phi_m^2 = \frac{n_p n_s}{n_i} \frac{\lambda_p \lambda_i}{\lambda_s L} , \qquad (1)$$

where

n_p , n_i , n_s are pump, idler, and signal refractive indices,

λ_p , λ_s , λ_i are pump, signal, and idler wavelengths,

L is crystal length, and

$\lambda_s > \lambda_i > \lambda_p$ will drive the backward wave process.

For the $LiTaO_3$ pumped with ruby this allowable divergence is about 0.5 milliradians. ($L = 2$ cm , $\lambda_s = 0.55$ mm) . Consider a laser having area A_p and radiating uniformly into an angle ϕ_p , where ϕ_p is greater than ϕ_m given by Eq. (1). By geometric optics any attempt to further focus the pump will lead to an increase in divergence. Since its divergence is already greater than ϕ_m , no further focusing of the pump is possible and only a fraction, ϕ_m^2/ϕ_p^2 , of the total power density is useful in driving the backward wave process.

The density is therefore given by

$$\left(\frac{P_p}{A}\right)_{max} = \frac{\varphi_m^2}{\varphi_p^2}\frac{P_p}{A_p}$$

$$= \frac{n_p n_s}{n_i}\frac{\lambda_p \lambda_i}{\lambda_s L}\frac{1}{\varphi_p^2 A_p} P_p \quad .$$

Hence 70 Mw/cm^2 radiating into 10 milliradians is equivalent to about 100 kw/cm^2 radiating within ϕ_m . Considerable effort has gone into transverse mode control as a means of reducing laser beam divergence. The use of a curved dielectric end mirror and a carefully selected aperature stop has provided us with a laser which operates with most of its energy in the lowest order (Gaussian) transverse cavity mode. Far field beam divergence has been reduced to less than 0.4 milliradians. Output spot size is about 10^{-2} cm^2 and output power is greater than 1 Mw.

With the laser pump essentially in the lowest order transverse mode the highest allowable focusing is dictated by the requirement that the signal, idler, and pump beams overlap in the crystal. This limits focusing to Rayleigh range at the far infrared wavelength $(\lambda_s L/2n_s)$ or approximately 10^{-2} cm^2. Hence the greatest available pumping density is approximately 100 Mw/cm^2 or more than eight times threshold for a non-cavity backward wave oscillator.

As a first step toward cavity free oscillation in the far infrared we propose to resonate the idler, thus only requiring gain to overcome cavity losses. Calculated threshold for round trip cavity loss of 15% is approximately 1.2 Mw/cm^2. See Fig. 11 for a schematic outline of the proposed experiment.

If single cavity backward wave oscillation is experimentally successful we will measure parametric gain by insertion of loss into the idler cavity just sufficient to quench the oscillation. If measured parametric gain is sufficient for cavity free backward wave oscillation the idler cavity will be removed and the cavity free experiment will be attempted.

Previous birefringence versus temperature measurements show that tuning should be possible from below the lowest Raman mode (135 microns) to a wavelength greater than 1 mm. See Fig. 12.

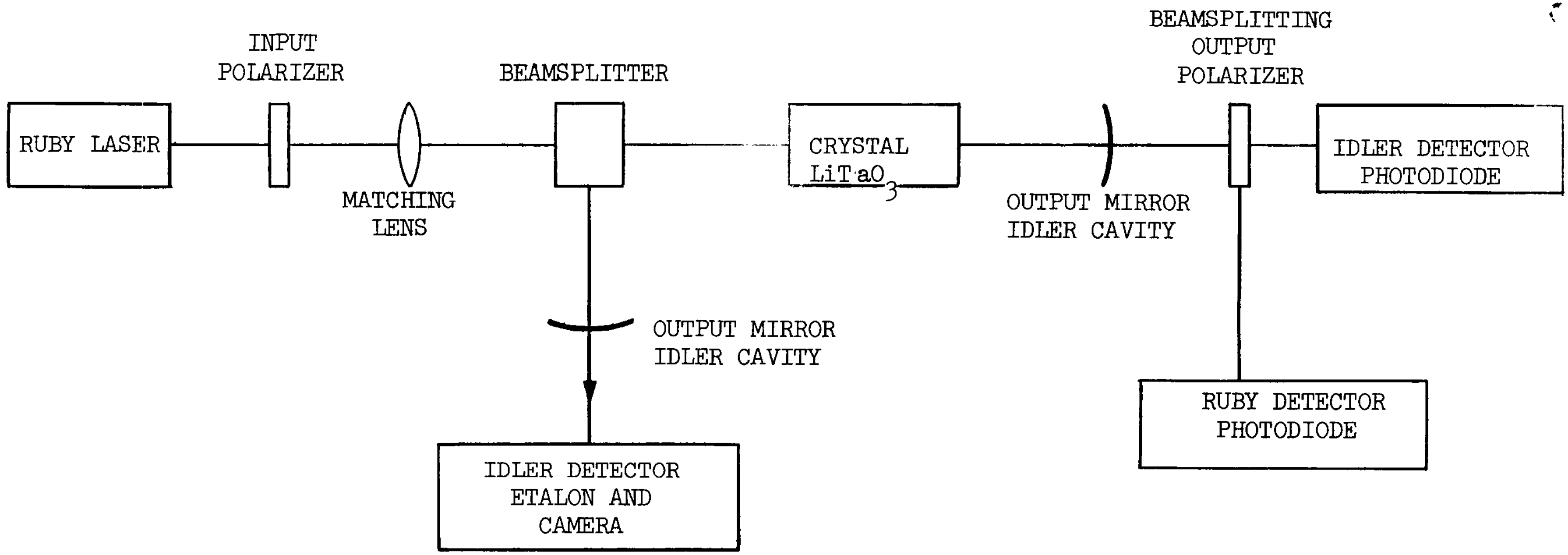

FIG. 11--Proposed backward wave oscillator experiment.

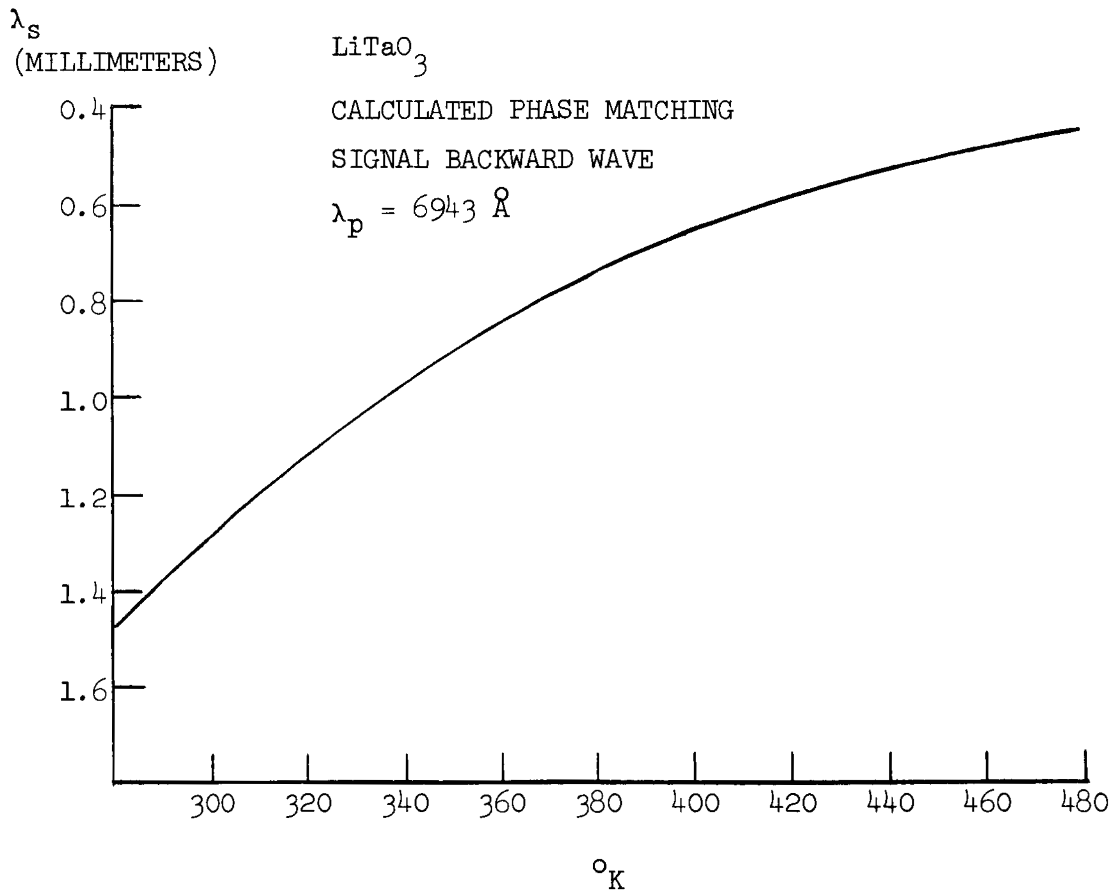

FIG. 12--Calculated phase matched signal wavelength, $LiTaO_3$ pumped with ruby. This curve is based on a 6328 Å measurement of birefringence versus temperature.

Although previous problems of poling $LiTaO_3$ have been solved, some difficulty remains in the aquisition of good optical quality crystals. A problem is anticipated in acquiring crystals whose birefringence does not wander along the length. This effect has been found to be severe in $LiNbO_3$ and is expected in $LiTaO_3$.